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Temperature dependence of the upper critical field H_{c2} of superconductors with inequivalent conducting layers

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Abstract : The temperature dependence of the upper critical magnetic field H_{c2} of high-T_c superconductors with inequivalent conducting layers has been studied using a Lawrence-Doniach (LD) type free energy functional suggested by Bulaevskii and Vagner [1]. At low temperatures ($T << T_c$), $H_{c2}^{\perp}(T)$ is a straight line with negative slope, but shows a positive curvature near T_c. The mass anisotropy in the ab-plane of the CuO chain layers in Y-Ba-Cu-O compounds enhances the curvature. $H_{c2}^{\parallel}(T)$ also shows positive curvature near T_c. The results are in general agreement with the experimental data for Y-Ba-Cu-O single crystals.

Keywords : High temperature superconductors, Lawrence-Doniach model, critical magnetic fields.

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1. Introduction

DC-magnetization measurements on single crystals of $YBa_2Cu_3O_7$ [1,2] of the upper critical field has established the positive curvature of H_{c2} near T_c for all field orientations. Such curvature has also been observed in low-T_c layered superconductors [3,4]. At lower temperatures $H_{c2}(T)$ becomes a straight line with negative slope intercepting the T-axis at about 1K below T_c [2]. The Ginzburg-Landau (GL) theory predicts linear temperature dependence for H_{c2} for all field orientations near T_c. The Lawrence-Doniach (LD) model of identical Josephson coupled layers [5], on the other hand, predicts a positive curvature near T_c for the upper critical field parallel to the layers ($H_{c2}^{\parallel}(T)$). But in this model $H_{c2}^{\parallel}(T)$ diverges at low temperatures. The positive curvature of $H_{c2}^{\perp}(T)$ however is not explained in this model.

The metallic nature of the CuO chains and the importance of the chain-plane interactions on the superconducting behaviour of $YBa_2Cu_3O_7$ has been well established [6–10]. Pathbreaking experiments by Kleiner *et al* [11] on the intrinsic Josephson effects on single crystals of YBCO, BSCCO and TBCCO strongly support a model of cuprate superconductors as a stack of superconducting sheets (S) consisting of CuO_2 layers (CuO_2 bilayers in the case of YBCO) separated by weakly superconducting (S') metallic layers (chain layers in YBCO). According to Kleiner *et al* superconducting S sheets induce a finite order parameter in the S' sheets which have no intrinsic superconductivity through a proximity effect and their experimental results support an $S-S'$ structure for YBCO with different order parameters for the S and S' sheets. Proximity effect has also been observed in Bi 2 : 2 : 1 : 2 by Briceno and Zettl. Based on these observations, it is possible to write a modified LD free energy functional for high-T_c superconductors on the lines suggested earlier by Bulaevskii and Vagner [1]. We have calculated elsewhere the fluctuation specific heat, paraconductivity and fluctuation contribution to the London penetration depth of high-T_c cuprates using such a model [15,16]. Other authors [13] have also considered the proximity effect between the planes and the chain layers, but introduced two coupling coefficients, one between the multiple CuO_2 planes and another between the planes and the chain layers. However experiments by Kleiner *et al* [11] clearly show that the adjacent multiple CuO_2 layers can be equated to a single superconducting sheet S and that it is not necessary to consider two coupling coefficients. Instead it is sufficient to consider the $S-S'$ coupling arising due to proximity effect. The scenario is the same in bismuth and thallium based superconductors also as they contain superconducting multiple CuO_2 layers separated by non-superconducting metallic layers (BiO and TlO layers respectively). Like the CuO chain layers in the yttrium compounds the metallic layers in these compounds act as charge reservoirs, dope charges into the CuO_2 layers and enhance the interlayer coupling [10,17–19].

In the present paper, we have studied the temperature dependence of the upper critical field H_{c2} both perpendicular and parallel to the layers starting with the modified LD free energy functional proposed by Bulaevskii and Vagner [1].

2. Upper critical field perpendicular to the layers

The free energy expression considered in ref. [8] is

$$
\begin{aligned}
G = \sum_n \int \Bigg[& a_1 \left|\psi_{1,n}^{(\rho)}\right|^2 + \frac{b_1}{2}\left|\psi_{1,n}^{(\rho)}\right|^4 + \frac{\hbar^2}{2m_{\|}}\left|\left(\nabla_{\|} - \frac{2ie}{\hbar c}A_{\rho,n}\right)\psi_{1,n}^{(\rho)}\right|^2 \\
& + a_2\left|\psi_{2,n}^{(\rho)}\right|^2 + \frac{b_2}{2}\left|\psi_{2,n}^{(\rho)}\right|^4 + \frac{\hbar^2}{2}\sum_{l=x,y}\frac{1}{m_l'}\left|\left(\frac{\partial}{\partial l} - \frac{2ie}{\hbar c}A_{l,n}\right)\psi_{2,n}^{(\rho)}\right|^2 \\
& + t\left|\psi_{1,n}^{(\rho)} - \psi_{2,n}^{(\rho)}e^{i\chi_n}\right|^2 + t\left|\psi_{1,n}^{(\rho)} - \psi_{2,n+1}^{(\rho)}e^{-i\chi_n}\right|^2\Bigg] d\rho \\
& + \int\left[\frac{h^2}{8\pi} - \frac{Hh}{4\pi} + \frac{H^2}{4\pi}\right] d\rho dz. \qquad (1)
\end{aligned}
$$

$\psi_{j,n}^{(\rho)}$ are the order parameters for layers $j = 1, 2$ in the unit cells numbered by the index n. The subscript 1 refers to the multiple CuO_2 layers and 2 to the metallic layers. In layer 2 we have taken into account the anisotropy of the effective mass due to the chain structure. $\rho = (x, y)$ and z is the axis perpendicular to the layers. $a_1(T) = \alpha_1 \dfrac{(T - T_0)}{T_c}$, $a_2(T) = \alpha_2$ and $\chi_n = \dfrac{2ed}{\hbar c} A_{z,n}$. α_1 and α_2 are positive constants. T_0 is some phenomenological temperature.

Near H_{c2}, the order parameters will have small values and hence the quartic terms in (1) can be neglected. Also $h = H$ and the free energy expression becomes,

$$G = \sum \int \Bigg[a_1 \left| \psi_{1,n}^{(\rho)} \right|^2 + \frac{\hbar^2}{2m_{\parallel}} \left| \left(\nabla_{\parallel} - \frac{2ie}{\hbar c} A_{\rho,n} \right) \psi_{1,n}^{(\rho)} \right.$$

$$+ a_2 \psi^{(\rho)} \qquad \frac{\hbar^2}{} \sum_{l=x,y} \frac{1}{m_l'} \quad \frac{\upsilon}{\partial l} - \frac{2ie}{\hbar c} A_{l,n} \Big| \psi_{2,n}^{(\rho)}$$

$$+ t \left| \psi_{1,n}^{(\rho)} - \psi_{2,n}^{(\rho)} e^{i\chi_n} \right|^2 + t \left| \psi_{1,n}^{(\rho)} - \psi_{2,n+1}^{(\rho)} e^{-i\chi_n} \right|^2 \Bigg] d\rho. \tag{2}$$

We shall assume the magnetic field on each layer as

$$H_n = H(\hat{z} \cos\theta + \hat{x} \sin\theta \cos\phi + \hat{y} \sin\theta \sin\phi). \tag{3}$$

Then the vector potential

$$A_n = H\left[-\hat{x} \frac{y \cos\theta}{2} + \hat{y} \frac{x \cos\theta}{} + \hat{z}(-x \sin\theta \sin\phi + y \sin\theta \cos\phi) \right] \tag{4}$$

and $$\chi_n = 2Kd \sin\theta(-x \sin\phi + y \cos\phi)$$

where $K = eH/\hbar c$. When the magnetic field is applied perpendicular to the layers, $\theta = 0$,

$$H_n = H\hat{z}, \qquad A_n = \frac{H}{2}(-y\hat{x} + x\hat{y})$$

and $\chi_n = 0$. The free energy (2) is minimized with respect to the variations in $\psi_{1,n}^*$ and $\psi_{2,n}^*$ and the real and imaginary parts of the resulting LD equations are separately equated to zero to obtain the following equations.

$$a_1 \psi_{1,n}^{(\rho)} - \frac{\hbar^2}{2m_1} \frac{\partial^2 \psi_{1,n}^{(\rho)}}{\partial x^2} + \frac{\partial^2 \psi_{1,n}^{(\rho)}}{\partial y^2} - K^2(x^2 + y^2)\psi_{1,n}^{(\rho)}$$

$$+ t\left[2\psi_{1,n}^{(\rho)} - \psi_{2,n}^{(\rho)} - \psi_{2,n+1}^{(\rho)} \right] = 0 \tag{5a}$$

$$y \frac{\partial \psi_{1,n}}{\partial x} = x \frac{\partial \psi_{1,n}}{\partial y}$$

$$a_1\psi_{2,n}^{(\rho)} - \frac{\hbar^2}{2m_{2x}}\frac{\partial^2\psi_{2,n}^{(\rho)}}{\partial x^2} - \frac{\hbar^2}{2m_{2x}}\frac{\partial^2\psi_{2,n}^{(\rho)}}{\partial y^2} - \frac{\hbar^2K^2}{2}\left(\frac{x^2}{m_{2y}}+\frac{y^2}{m_{2x}}\right)\psi_{2,n}^{(\rho)}$$

$$+t\left[2\psi_{2,n}^{(\rho)} - \psi_{1,n}^{(\rho)} - \psi_{1,n-1}^{(\rho)}\right] = 0 \tag{5b}$$

and $$\frac{y}{m_{2x}}\frac{\partial\psi_{2,n}}{\partial x} = \frac{x}{m_{2y}}\frac{\partial\psi_{2,n}}{\partial y}.$$

$m_1 = m_{\|}$. We sum eqs. (5) over all n, defining $\psi_j = \sum_n \psi_{j,n}/\sqrt{N}$ for $j = 1, 2$ where N is the number of layers considered. The equations for ψ_1 and ψ_2 which have the form

$$a_1\psi_1 - \frac{\hbar^2}{2m_1}\left[\frac{\partial^2\psi_1}{\partial x^2}+\frac{\partial^2\psi_1}{\partial y^2} - K^2(x^2+y^2)\psi_1\right] + 2t\left[\psi_1 - \psi_2\right] = 0 \tag{6a}$$

$$a_1\psi_2 - \frac{\hbar^2}{2m_{2x}}\frac{\partial^2\psi_2}{\partial x^2} - \frac{\hbar^2}{2m_{2y}}\frac{\partial^2\psi_2}{\partial y^2} - \frac{\hbar^2K^2}{2}\left(\frac{x^2}{m_{2y}}+\frac{y^2}{m_{2x}}\right)\psi_2$$

$$+2t\left[\psi_2 - \psi_1\right] = 0 \tag{6b}$$

Reconstructing an effective free energy functional from eqs. (6a) and (6b), we get,

$$\mathcal{F}_{\text{eff}} = \int\left[a_1|\psi_1|^2 + \frac{\hbar^2}{2m_1}\left|\frac{\partial\psi_1}{\partial x}\right|^2 + \left|\frac{\partial\psi_1}{\partial y}\right|^2 + \frac{\hbar^2K^2}{2m_1^2}(x^2+y^2)|\psi_1|^2\right.$$

$$+a_2|\psi_2|^2 + \frac{\hbar^2}{2m_{2x}}\left|\frac{\partial\psi_2}{\partial x}\right|^2 + \frac{\hbar^2}{2m_{2y}}\left|\frac{\partial\psi_2}{\partial y}\right|^2 + \frac{\hbar^2K^2}{2}\left(\frac{x^2}{m_{2y}}+\frac{y^2}{m_{2x}}\right)|\psi_2|^2$$

$$\left.+2t|\psi_1 - \psi_2|^2\right]dxdy \tag{7}$$

Let us put $x = y = 0$ and $T = T_c$ in eqs. (6). Near T_c, ψ_1 and ψ_2 will be very small and hence the spatial variations of these functions can be neglected. Therefore we get,

$$a_1(T_c)\psi_1 + 2t\psi_1 - 2t\psi_2 = 0 \tag{8a}$$

and $$a_2(T_c)\psi_2 + 2t\psi_2 - 2t\psi_1 = 0 \tag{8b}$$

Eqs. (8) have nontrivial solutions only when

$$a_1(T_c) + 2t - \frac{4t^2}{a_2+2t} = 0 \tag{9a}$$

Let us assume $a_1(T) = \alpha_1(T - T_0)/T_c$ and $a_2(T) = \alpha_2$ where T_0 is some phenomenological temperature and α_1 and α_2 are positive constants. Here it is assumed that the temperature dependence appears only in the coefficient associated with the CuO_2 planes. From (9a), we obtain the relation

$$\frac{T_0}{T_c} = 1 + \frac{2t}{\alpha_1} - \frac{4t^2}{\alpha_1(\alpha_2 + 2t)}. \tag{9b}$$

The free energy functional (7) may be written in terms of

$$\lambda = \frac{4t^2}{\alpha_1(\alpha_2 + 2t)}, \qquad \xi^2 = \frac{\hbar^2}{2m_1\alpha_1},$$

$$\ell_x^2 = \frac{\lambda^2 \hbar^2 \alpha_1}{8m_{2x}t^2}, \qquad \ell_y^2 = \frac{\lambda^2 \hbar^2 \alpha_1}{8m_{2y}t^2}.$$

and the rescaled order parameters $\psi_1' = \alpha_1 \psi_1$ and $\psi_2' = \frac{2t}{\lambda}\psi_2$. After dropping the primes for convenience the effective free energy can be written as

$$G_{\text{eff}} = \int \Bigg[\left(\frac{T}{T_c} - 1\right)|\psi_1|^2 + \xi^2 \frac{\partial \psi_1}{\partial x} \frac{\partial \psi_1}{\partial y} + \xi^2 K^2 (x^2 + y^2)|\psi_1|^2$$

$$+ \ell_x^2 \frac{\partial \psi_2}{\partial x} + \ell_y^2 \frac{\partial \psi_2}{\partial y} K^2 \left(\ell_y^2 x^2 + \ell_x^2 y^2\right)|\psi_2|^2$$

$$+ \lambda |\psi_1 - \psi_2|^2 \Bigg] dxdy \tag{10}$$

The equations that minimize G_{eff} are

$$\xi^2 \left(\frac{\partial^2 \psi_1}{\partial x^2} + \frac{\partial^2 \psi_1}{\partial y^2}\right) = \xi^2 K^2 (x^2 + y^2)\psi_1 + \left(\frac{T}{T_c} - 1\right)\psi_1 + \lambda[\psi_1 - \psi_2] \tag{11a}$$

and

$$\ell_x^2 \frac{\partial^2 \psi_2}{\partial x^2} + \ell_y^2 \frac{\partial^2 \psi_2}{\partial y^2} = K^2 \left(\ell_y^2 x^2 + \ell_x^2 y^2\right)\psi_2 + \lambda[\psi_2 - \psi_1] \tag{11b}$$

The solutions to eqs. (11a) and (11b) in the limit $\lambda \to 0$ and $T \to T_c$ can be written as

$$\psi_1 = \psi_{10} e^{-\gamma(x^2 + y^2)}$$

and

$$\psi_2 = \psi_{20} e^{-\gamma\left(\frac{\ell_y}{\ell_x} x^2 + \frac{\ell_x}{\ell_y} y^2\right)} \tag{12}$$

where $\gamma = \frac{K}{2}$. Substituting (12) back into (11), we get,

$$\left(\frac{T}{T_c} - 1 + \lambda + 2K\xi^2\right)\psi_{10} - \lambda\psi_{20} = 0 \tag{13}$$

and

$$\lambda\psi_{10} - \left(\lambda + 2K\ell^2\right)\psi_{20} = 0 \tag{14}$$

where $\ell^2 = \ell_x \ell_y$. Eqs. (13) and (14) have nontrivial solutions only if

$$1 - \frac{T}{T_c} = \lambda + 2K\xi^2 - \frac{\lambda^2}{(2K\ell^2 + \lambda)} \tag{15}$$

Eq. (15) determines the temperature dependence of $H_{c2}^{\perp}$.

As $H \to \infty$, $\quad 1-\frac{T}{T_c} = \lambda + 2K\xi^2.$

At large H-values, $H_{c2}^{\perp}(T)$ becomes a straight line which cuts the T-axis at $T = T_c(1-\lambda)$. In SI units $H_{c2}^{\perp}(T)$ can be written as

$$H_{c2}^{\perp}(T) = \frac{(1-\lambda)\hbar}{2e\xi^2} - \frac{1}{T_c}\frac{\hbar}{2e\xi^2}T. \tag{16}$$

Comparing this with the linear fit for critical field data on single crystal $YBa_2Cu_3O_7$ for fields perpendicular to the layers given in ref. [2]

$$H_{c2}^{\perp} = 173.1689 - 1.8919\,T$$

we obtain $\xi = 13.725$ Å and $\lambda = 0.0098$. For small values of H,

$$\frac{dH}{dT} = -\frac{1}{T_c}\frac{\hbar c}{2e}\frac{1}{L^2}$$

where $L^2 = \xi^2 + \ell^2$. Low field critical field data on $YBa_2Cu_3O_7$ single crystals [14] yields $\frac{dH}{dT} = 80$ G/K. This is smaller than the critical field slope observed at higher fields by a factor of 100. From the expression for $\frac{dH}{dT}$ we get $\ell = 210.5$ Å. The curvature

$$\frac{1}{R} = \frac{\hbar c}{2e}\frac{2\ell^2}{\lambda T_c^2 L^4\left[1+\frac{\hbar^2 c^2}{4e^2T_c^2}L^{-4}\right]}$$

is positive. Experimental data on single crystal $YBa_2Cu_3O_7$ [2,14] show that the curvature is present for H-values upto 18 G, depends on the mass anisotropy in the CuO chain layers and is enhanced by it.

Reliable H_{c2} data is also available for the $La_{1.87}Ca_{1.13}Cu_2O_6$ [20] for which the double CuO_2 planes in the crystallographic unit cell constitutes the S layer and the double metallic LaO planes from the S' layer. Our calculations based on the experimental data in ref. [20] give $\lambda = 0.1694$, $\xi = 30.1$ Å and $\ell = 233$ Å for this compound. This value of ξ compares favourably with the experimental value of 33 Å. The fairly strong coupling coefficient obtained from the calculations agrees with the rather small anisotropy of the coherence length which implies a fairly isotropic electronic structure for the compound inspite of its quasi two dimensional crystal structure.

From eq. (14),

$$\frac{\psi_{20}}{\psi_{10}} = \frac{\lambda}{\lambda + 2K\ell^2}. \tag{17}$$

As

$$H \to 0, \quad \frac{\psi_{20}}{\psi_{10}} \to 1.$$

But as H increases $\frac{\psi_{20}}{\psi_{10}} \to 0$ (see Figure 1). Therefore the linear region of $H_{c2}^{\perp}(T)$ is precisely the region where the order parameter becomes zero on the S' layers.

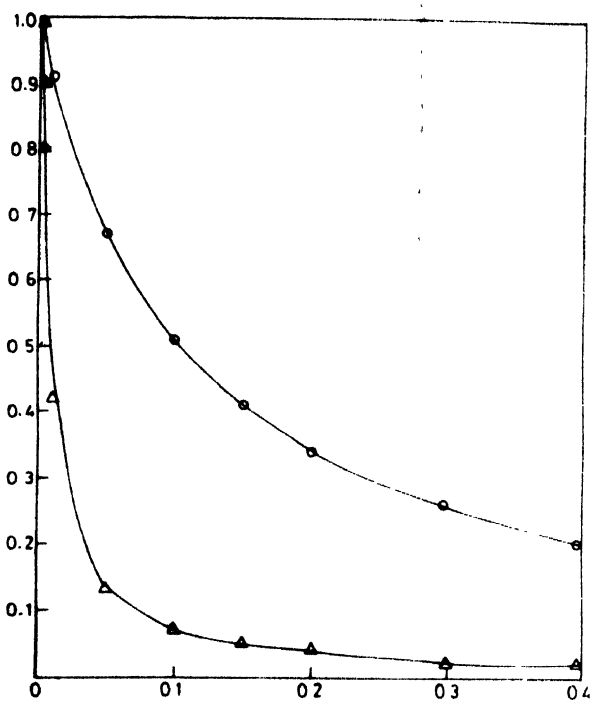

Figure 1. The ratio of the NSC to the SC order parameter for fields parallel to the C-axis. Triangles represent $YBa_2Cu_3O_7$ and circles represent $La_{1.87}Ca_{1.13}Cu_2O_6$

The temperature dependence of $H_{c2}^{\perp}$ can also be obtained by a variational method. We try the solution

$$\psi_1 = be^{-\gamma(x^2+y^2)}$$

and $$\psi_2 = e^{-\gamma(x^2+y^2)} \tag{18}$$

where b and γ are variational parameters. We now make use of the fact that G_{eff} given by (10) should vanish at the transition from the superconducting to the normal state. Substituting eq. (18) in (10), performing the integration with the help of standard integrals [21] and setting $G_{\text{eff}} = 0$, we get

$$\left[\left(\frac{T}{T_c} - 1 + \lambda\right)\gamma + 2\xi^2 g_1\right] b^2 - 2b\lambda\gamma + \left[\left(r_x^2 + r_y^2\right)g_1 + \lambda\gamma\right] = 0 \tag{19}$$

where $g_1 = \gamma^2 + \frac{K^2}{4}$. Maximizing $T_c(H)$ given by eq. (19) with respect to b, we obtain

$$-\frac{T}{T_c} = \lambda + 2\xi^2 g_1 - \frac{\lambda^2}{\left[\left(\ell_x^2 + \ell_y^2\right)g_1 + \lambda\right]}$$

Since λ is small, $T_c(H)$ can be maximized with respect to γ neglecting the term in λ^2. Then we get $\gamma = \frac{K}{2}$ and

$$1 - \frac{T}{T_c} = \lambda + 2\xi^2 K - \frac{\lambda^2}{\left[\left(\ell_x^2 + \ell_y^2\right)K + \lambda\right]} \tag{20}$$

Eq. (20) is similar to eq. (15) and validates all the features of the $H_{c2}^{\perp}(T)$ discussed earlier. In the case of thallium and bismuth based compounds and the 214 compound S' layers are square planar. Therefore $\ell_x = \ell_y$ and eqs. (15) and (20) are identical. In the LD case of identical superconducting planes, $\psi_2 = 0$. If we set $\lambda = 0$,

$$\psi_1 = \psi_{10} e^{-\frac{K}{2}(x^2+y^2)}.$$

The temperature dependence of the upper critical field in this case is obtained from eq. (13) as

$$1 - \frac{T}{T_c} = 2K\xi^2.$$

For small λ, the correction to H_{c2} can be obtained by evaluating G_{eff} with the above unperturbed functional forms for ψ_1 and ψ_2 and equating the result to zero. This yields

$$1 - \frac{T}{T_c} = \lambda + 2K\xi^2. \tag{21}$$

When $H = 0$, $T \neq T_c$ and hence this result is not valid near T_c. However it agrees with the large H-limit of eq. (15). The situation here is identical to the LD model with equidistant identical superconducting layers.

Another possible model for $YBa_2Cu_3O_7$ with inequivalent conducting layers [22] is obtained by ignoring the proximity effect and the contribution of the S' layers and considering the two CuO_2 layers in the elementary cell separately. The distance between the CuO_2 layers in the same elementary cell is $d_1 = 3.3$ Å and that between layers in the neighbouring cells is $d_2 = 8.4$ Å. This entails the introduction of two different coupling coefficients t_1 and t_2 and two different order parameters ψ_1 and ψ_2 to represent the inequivalently placed CuO_2 layers. The Gibb's free energy for this model is

$$G = d\sum_n \int\left[a\left(|\psi_{1,n}|^2 + |\psi_{2,n}|^2\right) + \frac{b}{2}\left(|\psi_{1,n}|^4 + |\psi_{2,n}\right.\right.$$

$$\left.\left. + \frac{\hbar^2}{2m}\left|\left(\nabla_{\parallel} - \frac{2ie}{\hbar c}A_{\parallel}\right)\psi_{1,n}\right|^2 + \frac{\hbar^2}{2m}\left|\left(\nabla_{\parallel} - \frac{2ie}{\hbar c}A_{\parallel}\right)\psi_{2,n}\right.\right.$$

$$+t_1\left|\psi_{1,n}-\psi_{2,n}e^{-i\frac{2ed_1}{\hbar c}A_{zn}}\right|^2+t_2\left|\psi_{1,n}-\psi_{2,n-1}e^{-i\frac{2ed_2}{\hbar c}A_{zn}}\right|^2\Bigg]d\rho$$

$$\left[\frac{h^2}{8\pi}-\frac{h.H}{4\pi}+\frac{H^2}{4\pi}\right]d\rho dz \tag{22}$$

where $d = d_1 + d_2$, $a_1 = a_2 = a$, and $m_{2x} = m_{2y} = m_1 = m$. Proceeding as before, we find :

$$1-\frac{T}{} = 2\ell_1^2K \tag{23}$$

The $H_{c2}^{\perp}(T)$ graph is a straight line which cuts the T-axis at $T = T_c$ and there is no curvature for low values of H. Comparison of the results (21) and (23) show conclusively that the positive curvature which is a generic property of high-T_c superconductors can be explained only if the contribution from the S' layers which are structurally different from the S layers are also included in the free energy expression. The inequivalency of the CuO_2 layers as described by (22) alone does not suffice to explain the positive curvature of $H_{c2}^{\perp}(T)$.

3. Angular dependence of H_{c2}

When the magnetic field is applied in the x-z plane, $\phi = 0$,

$$H_n = H(\hat{z}\cos\theta + \hat{x}\sin\theta)$$

and

$$A_n = yH(\hat{z}\sin\theta - \hat{x}\cos\theta).$$

In this case the order parameters have no explicit x-dependence and proceeding as in section 2, we get

$$G_{\text{eff}} = \int\Bigg[\left(\frac{T}{T_c}-1\right)|\psi_1|^2+\xi^2\left|\frac{\partial\psi_1}{\partial y}\right|^2+4\xi^2K^2\cos^2\theta y^2|\psi_1|+\ell_y^2\frac{\partial\psi_2}{\partial y}$$

$$+4K^2\ell_x^2y^2\cos^2\theta|\psi_2|^2+\frac{\lambda}{2}|\psi_1-\psi_2\exp(i2Kdy\sin\theta)|^2$$

$$+\frac{\lambda}{2}|\psi_1-\psi_2\exp(-i2Kdy\sin\theta)|^2\Bigg]dy$$

Using trial solutions

$$\psi_1 = b\exp(-\gamma y^2) \quad \text{and} \quad \psi_2 = \exp(-\gamma y^2),$$

G_{eff} is evaluated with the help of standard integrals [21] and it is set equal to zero at the transition from the superconducting to the normal state *i.e.* when $H = H_{c2}$. In the resulting expression $T_c(H)$ is maximized with respect to b and we get

$$1-\frac{T}{T_c} = \lambda+\xi^2g_2-\frac{\lambda^2\exp\left(-\dfrac{e^2d^2H^2\sin^2\theta}{\hbar^2c^2\gamma}\right)}{\left[\ell_y^2\gamma+\ell_x^2\dfrac{K^2\cos^2\theta}{\gamma}+\lambda\right]} \tag{24}$$

where $g_2 = \gamma + \frac{K^2 \cos^2\theta}{\gamma}$. In the general case maximization of this expression with respect to γ can be done only numerically. However for large fields when $\left(\ell_y^2\gamma + \ell_x^2 \frac{K^2\cos^2\theta}{\gamma}\right) > \lambda$, the last term in λ^2 becomes negligible and $\gamma = K\cos\theta$. In the large H limit, the temperature and angular dependence of H_{c2} is given by

$$1 - \frac{T}{T_c} = \lambda + 2K\xi^2\cos\theta - \frac{\lambda^2}{\left(\ell_x^2 + \ell_y^2\right)K\cos\theta}\exp\left(-\frac{Kd^2\sin^2\theta}{\cos\theta}\right). \tag{25}$$

For $\theta = 0$, eq. (25) reduces to eq. (20). In the opposite limit, when $\lambda > \left(\ell_y^2\gamma + \ell_x^2\frac{K^2}{4\gamma}\right)$ the exponential in eq. (24) is expanded to the first order in the argument and then maximized with respect to γ to obtain.

$$\gamma = \frac{eH}{\hbar c}\left(\cos^2\theta + \varepsilon\sin^2\theta\right)^{\frac{1}{2}} \tag{26}$$

where $\varepsilon = \lambda d^2/\xi^2$. In this limit, the temperature and angular dependence of H_{c2} is obtained as

$$1 - \frac{T}{T_c} = \frac{2eH\xi^2}{\hbar c}\left[\cos^2\theta - \varepsilon\sin^2\theta\right]^{\frac{1}{2}}. \tag{27}$$

Eq. (27) is the θ-dependence obtained from the GL anisotropic theory and was experimentally verified in ref. [23].

4. Upper critical field parallel to the layers

Our results (25) and (27) cannot be relied upon for $\theta = 90°$ since in this case the order parameters are oscillatory Mathieu functions as we will see later in this section. Setting $\theta = 90°$, eq. (27) yields

$$1 - \frac{T}{T_c} = 2\sqrt{\lambda}\,\frac{ed\xi}{\hbar c}H,$$

which represents a straight line with negative slope passing through $T = T_c$. We can also calculate the temperature dependence of $H_{c2}^{\parallel}$ by the variational method. G_{eff} given by (10a) is evaluated after setting $\theta = 90°$, using the ansatz (18a) and is set equal to zero. This gives us $T_c(H)$ as a function of variational parameters. The maximization of $T_c(H)$ with respect to b can be done analytically whereas the same with respect to γ can be done only in certain limiting cases. In the limit $\lambda > \ell_y^2\gamma$,

$$1 - \frac{T}{T_c} = \lambda + \frac{\xi^2 dK}{L_y} - \frac{\lambda^2(1 - L_y dK)}{\left(\frac{\ell_y^2 dK}{L_y} + \lambda\right)} \tag{28}$$

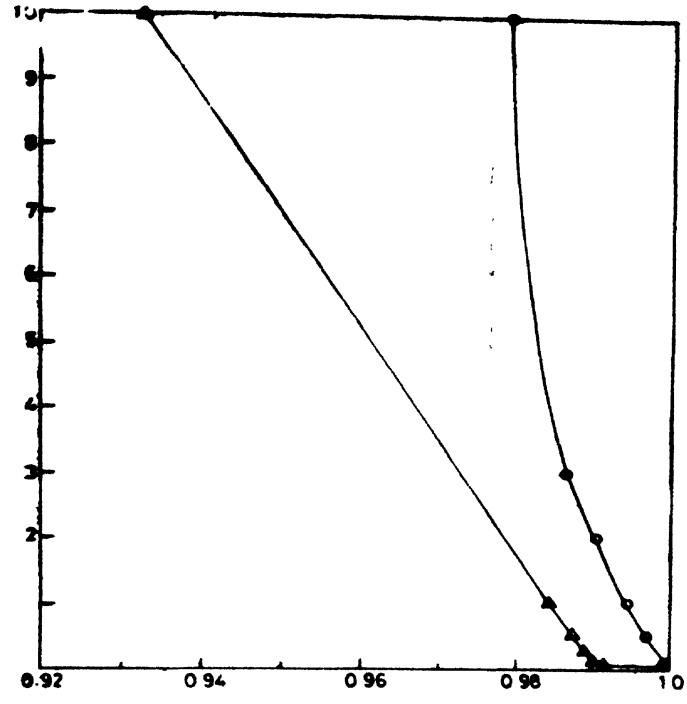

Figure 2. Graphical representation of $H_{c2}^{\perp}(T)$ and $H_{c2}^{\parallel}(T)$ for $YBa_2Cu_3O_7$ as given by equations (20) and (28).

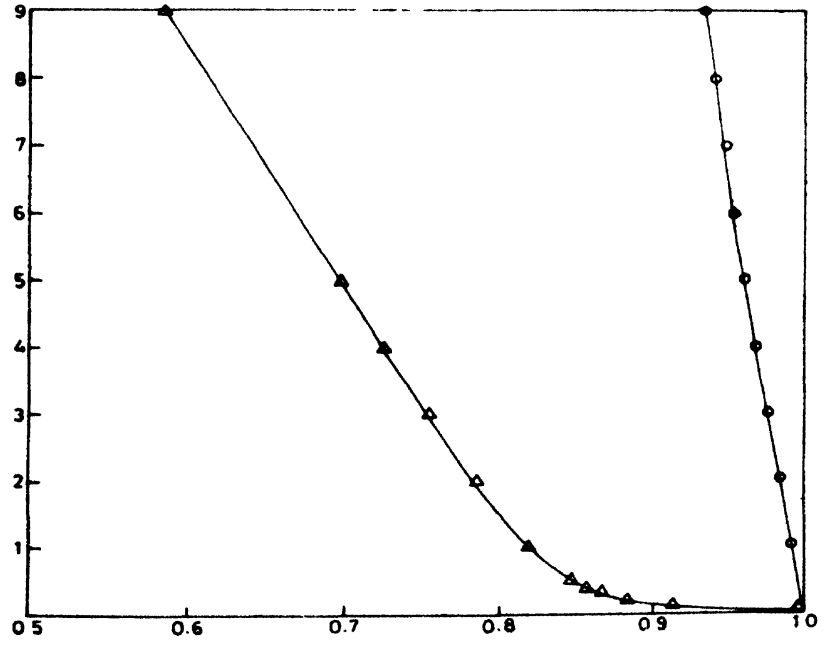

Figure 3. Graphical representation of $H_{c2}^{\perp}(T)$ and $H_{c2}^{\parallel}(T)$ for $La_{1.87}Ca_{1.13}Cu_2O_6$ as given by equations (20) and (28).

where $L_y^2 = (\xi^2 + \ell_y^2)/\lambda$. At large values of H, $H_{c2}^{\parallel}(T)$ is a straight line which on extrapolation cuts the T-axis at $T^* = T_c(1-2\lambda)$. It is interesting to consider the case when $\psi_2 = 0$ and only identical equispaced double CuO_2 planes are considered. The situation is identical to the LD formalism. In this case $T_0 = T_c$. The free energy expression for determining $H_{c2}^{\parallel}$ in this case becomes [24]

$$F_S = \sum_n \int dx \left[a|\psi_n|^2 + \frac{\hbar^2}{2m_{\parallel}} \left| \frac{\partial \psi_n}{\partial y} \right| + t|\psi_n - \psi_{n-1} e^{i\chi_n}|^2 \right]$$

$\psi_n(y) = \psi(y)\exp(-ikn)$ Differentiating F_S with respect to ψ^* we obtain the Mathieu equation

$$-\frac{\hbar^2 \partial^2 \psi}{2m_{\parallel} \partial y_2^2} + 2t\psi\left[1 - \cos\frac{2edH}{\hbar c} y\right] = -a\psi. \tag{29}$$

If $E(H)$ is the lowest eigen value of this equation, transition to the normal state occurs when $E(H) \geq |a|$. For small values of H the cosine term is expanded to the first order in the argument. We obtain the Schröedinger equation for the harmonic oscillator, solving which we get

$$H_{c2}^{\parallel} = \left(\frac{mc^2\alpha^2}{2e^2d^2t}\right)^{\frac{1}{2}} \frac{T - T_c}{T_c}. \tag{30}$$

At large H, $E(H) \geq |a| > t$. For $t < E < 2t$ eq. (29) can be solved to obtain

$$H_{c2}^{\parallel} = \left[\frac{mc^2t^2}{e^2d^2\alpha} \frac{T_c - T^*}{T_c}\right]^{\frac{1}{2}}, \tag{31}$$

where $T^* = T_c(1-2\lambda)$. When $|a| > 2t$, $E(H)$ is always smaller than $|a|$ and $H_{c2}^{\parallel}$ becomes infinite. Thus $H_{c2}^{\parallel}(T)$ exhibits positive curvature even if we consider a system of identical layers. This shows that while inclusion in the free energy of the contributions from the non superconducting layers arising from the proximity effect is essential for explaining the positive curvature of $H_{c2}^{\perp}(T)$, the LD model with its equidistant identical superconducting layers can also explain the positive curvature of $H_{c2}^{\parallel}(T)$.

The preceding procedure can be extended to $La_{2-x}Sr_xCuO_4$ superconductor with ψ_1 and ψ_2 representing the CuO_2 mono layers and LaO bilayers respectively and also to the thallium and bismuth based superconductors. Figure 2 is a graphical representation of eqs. (20) and (28) for $YBa_2Cu_3O_7$ compound whereas Figure 3 is the same for $La_{1.87}Ca_{1.13}Cu_2O_6$ superconductor. Eq. (28) is valid upto 2.6 T for $YBa_2Cu_3O_7$ and upto 37 T for $La_{1.87}Ca_{1.13}Cu_2O_6$.

5. Conclusion

We have theoretically studied the temperature dependence of the critical magnetic fields of $YBa_2Cu_3O_7$ superconductor by employing the modified LD free energy functional (1). For temperatures close to T_c, $H_{c2}^{\perp}(T)$ has a positive curvature and becomes a straight line with negative slope at large H values. The theoretical expression (16) was fitted to the experimental data for $YBa_2Cu_3O_7$ and $La_{1.87}Ca_{1.13}Cu_2O_4$. The values of the parameter ξ obtained in these two cases agree with the respective experimental values. $H_{c2}^{\parallel}(T)$ also exhibits positive curvature. Our treatment shows that the curvature depends not only on the inequivalency of the order parameters but also on the mass anisotropy of the CuO chain layers. Comparisons of models described by free energies (1) and (22) show conclusively that the non-zero value of the order parameter on the NSC layers which are structurally different from the SC layers is essential to explain the positive curvature of $H_{c2}^{\perp}(T)$ whereas the positive curvature of $H_{c2}^{\parallel}(T)$ can be explained even on the standard LD model. This is also borne out by the calculations in ref. [13] where the authors obtain a positive curvature for $H_{c2}^{\perp}$ considering the proximity effect between the CuO_2 planes and the CuO chain layers and by introducing two different coupling coefficients. The large H-limit of H_{c2} given by (16) and the parameters deduced therein are exactly identical to those of ref. [13] which is nothing but the expression (21) obtained for the LD model. However the experiments of Kleiner *et al* substantiates the simple model that we have considered in arriving at the results (15), (16), (20), (25), (26) and (28).

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